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Review

Spatial aspects of the dioxin risk formation in the Baltic Sea: A systematic review

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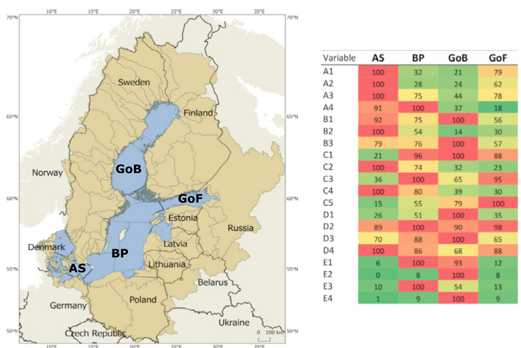
HIGHLIGHTS

- Comparable data on key variables of environmental health risk are compiled.
- The risk is a product of dioxin pollution, ecosystem dynamics, and fish consumption.
- Dioxin flux characteristics of the analyzed four Baltic Sea basins are diverse.
- Differences arise from the emissions, origin of fish, and consumption distinctions.
- The approach aid acknowledge spatial features of the dioxin risk and its management.

GRAPHICAL ABSTRACT



Variable	AS	BP	GoB	GoF
A1	100	32	21	79
A2	100	28	24	62
A3	100	75	44	78
A4	91	100	37	18
B1	92	75	100	56
B2	100	54	14	30
B3	79	76	100	57
C1	21	96	100	88
C2	100	74	32	23
C3	36	100	65	95
C4	100	80	39	30
C5	15	55	79	100
D1	26	51	100	35
D2	89	100	90	98
D3	70	88	100	65
D4	100	86	68	88
E1	6	100	93	12
E2	0	8	100	8
E3	10	100	54	13
E4	1	9	100	9

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ABSTRACT

Dioxins have been an inconvenience to the Baltic Sea ecosystem for decades. Although the concentrations in the environment and biota have continuously decreased, dioxins still pose a risk to human health. The risk and its formation vary in different parts of the Baltic Sea, due to variability in the environmental and societal factors affecting it. This paper presents a systematic literature review and knowledge synthesis about the regional dioxin risk formation in four sub-areas of the Baltic Sea and evaluates, whether systemic approach changes our thinking about the risk and its effective management. We studied the dioxin flux from atmospheric deposition to the Baltic Sea food webs, accumulation to two commercially and culturally important fish species, Baltic herring (*Clupea harengus membras*) and Baltic salmon (*Salmo salar*), and further to risk group members of four Baltic countries. Based on 46 studies, we identified 20 quantifiable variables and indexed them for commensurable regional comparison. Spatial differences in dioxin pollution, environmental conditions, food web dynamics, and the following dioxin concentrations in herring and salmon, together with fishing and fish consumption, affect how the final health risk builds up. In the southern Baltic Sea, atmospheric pollution levels are relatively high and environmental processes to decrease bioavailability of dioxins unfavorable, but the growth is fast, which curb the bioaccumulation of dioxins in the biota. In the North, long-range atmospheric pollution is minor compared to South, but the local pollution and slower growth leads to higher bioaccumulation rates. However, based on our results, the most remarkable differences in the dioxin risk formation between the areas arise from the social sphere: the emissions, origin of national catches, and cultural differences in fish consumption. The article suggests that acknowledging spatial characteristics of socio-ecological systems that generate environmental risks may aid to direct local focus in risk management.

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1. Introduction

Ecosystem-based management entails viewing complex problems as coupled social-ecological systems, identifying underlying factors, and accounting for their interactions and dynamics (Berkes, 2012; Long et al., 2015). The approach requires considering the most appropriate regional scale for analyses, in order to facilitate understanding and managing problems in holistic terms (Fletcher et al., 2010; Rosenberg et al., 2006). Yet, despite the increasing calls for ecosystem-based management, the management of complex problems is predominantly based on reductionist analyses focusing on specific parts of the problems, whereas holistic regional-level analyses are rare. This also applies to the dioxin problem of Baltic herring (*Clupea harengus membras*) and salmon (*Salmo salar*) fisheries.

Dioxins and dioxin-like compounds are persistent organic pollutants (POPs), usually referring to the most toxic and persistent 17 polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) and 12 dioxin-like polychlorinated biphenyls (dl-PCBs). They accumulate in the fatty tissue of organisms and hence in food chains. Thus, dioxins may cause adverse health effects on top predators, including humans (Kulkarni et al., 2008; White and Birnbaum, 2009). The amount of dioxins in an individual organism depends on species-specific physiological characteristics, the quantity and source of the emissions (Assefa et al., 2019), the surrounding environmental conditions (Armitage et al., 2009; Skei et al., 2000), and the food web interactions (Broman et al., 1992; Burreau et al., 2006; Nfon et al., 2008; Rolff et al., 1993; Vuorinen et al., 2012).

As fatty fish, Baltic herring and salmon generally accumulate dioxins from the food web more than fish with lower fat content. In general, the largest and oldest fish contain the highest dioxin levels (Airaksinen et al., 2014). However, due to differences in the area-specific characteristics, the amount of dioxins in fish varies in different parts of the Baltic Sea (Vuorinen et al., 2012). Human activities (e.g. fishing) and human-induced pressures (e.g. nutrient loading) may influence the trophic and species relationships in the food web, hence impacting the dioxin flux (Hoegh-Guldberg and Bruno, 2010).

Baltic herring and salmon have certain positive effects on human health, due to their relatively high concentrations of omega-3 fatty acids and vitamin D (Assmuth, 2011; Tuomisto et al., 2020, but see EMA, 2019). However, dioxins in Baltic herring and salmon offer argument against eating them. In earlier studies, high dioxin exposure has primarily been associated with enamel defects in teeth (Alaluusua et al., 1996; Alaluusua et al., 2004). Moreover, Mocarelli et al. (2008) first showed the evidence for the effect of TCDD exposure to decrease in sperm count and motility. Recently, evidence for the transfer of dioxin exposure in the germline of males who had been exposed to dioxins during nursing (Mocarelli et al., 2011) and prenatal periods (Mínguez-Alarcón et al., 2017; Mocarelli et al., 2011) was found. Correspondingly, the European Food Safety Authority (EFSA) (Knutsen et al., 2018) set a new, lower tolerable weekly intake (TWI, 2 pg kg^{-1} bodyweight) for the risk assessments, in 2018.

The dioxin risk of Baltic fish to human health is managed by global, EU-level, and regional policies aiming to reduce dioxin emissions, EU-policies restricting the selling of fish containing dioxins, and national eating recommendations (Haapasaari et al., 2019). These policies are based on the current understanding of the sources and properties of dioxins, as well as their pathways to the environment and to human food, and their effects on human health (Assmuth and Jalonen, 2005; COM, 2001/C322/02). In the EU, the maximum allowed dioxin level has been set for fish to be sold for food and feed (EU, 2016).

At the beginning of the 2000s, the EU called for improved knowledge of the transfer of dioxins from emissions to humans, and for integrated approaches to reduce dioxins both in the environment and food/feed (COM, 2001/C322/02; 2003/338). The calls were responded to by Tuomisto et al. (2003) who highlighted the need to develop methods that combine ecological and human risk assessment. Similarly, Assmuth and Jalonen (2005) identified a need for knowledge on the dioxin risk formation chain and a comprehensive perspective on the management of the dioxin problem in relation to risks. So far the Baltic Sea research has focused on 1) the atmospheric sources, deposition, and concentration of POPs in the sediment (Armitage et al., 2009; Assefa et al., 2014a; Assefa et al., 2018; Korhonen et al., 2013; Sobek et al., 2014; Sobek et al., 2015; Sundqvist et al., 2009; Wiberg et al., 2009), 2) transfer of POPs in herring and salmon food chains (Burreau et al., 2006; Nfon et al., 2008; Vuorinen et al., 2012), 3) concentration of POPs in fish in the different Baltic Sea basins (Airaksinen et al., 2014; Miller et al., 2013; Piskorska-Pliszczynska et al., 2012), and 4) human exposure to dioxins in Baltic fish (Kiljunen et al., 2007; Struciński et al., 2013). The governance and management of the dioxin problem of Baltic Sea fish has been examined by Pihlajamäki et al. (2018), Haapasaari et al. (2019) and Haapasaari et al. (Submitted manuscript).

This paper applies the principles of ecosystem-based management, compiling an integrated approach to understand the entire dioxin risk formation chain at the regional level. The aims of the paper are to

1) improve the holistic understanding of the transfer of dioxins from deposition to the ecosystem and humans, by comparing the system in different areas of the Baltic Sea, 2) take a systemic perspective to the risk analysis by combining ecological and human risk factors at the regional level, and finally, 3) discuss whether and how an integrated analysis conducted at the regional level could change our understanding of the dioxin risk in the Baltic Sea area and promote new management approaches.

The study consists of four phases. First, we identified and categorized key factors behind the dioxin risk in the Baltic Sea. Second, we selected four Baltic Sea basins as a meaningful spatial division for the analysis. Third, using the key words under the categories we conducted a systematic literature review, identifying 20 variables comparable under the selected spatial division. Fourth, we developed an index approach for comparing the relative significance of these variables for the dioxin risk formation between the four Baltic Sea areas. To summarize and visualize the key differences in the areal risk formation, the results are synthesized using a traffic light plot.

The paper is structured as follows. Section 2 describes the materials and methods used in the study. Section 3 describes results, which are further discussed in Section 4.

2. Materials and methods

2.1. Problem framing and study area division

The first phase of the analysis is a review of studies and policy documents that focus on the chemical, ecological, environmental, and human exposure aspects of the dioxin problem. The studies were partly discovered through recommendations from experts involved in the BONUS GOHERR-project (Integrated governance of Baltic herring and salmon stocks involving stakeholders, 2015–2018), that focused on the governance and management of the dioxin problem in herring and salmon. The dioxin risk formation was identified as follows:

The historical dioxin loading stored in the sediment and riverine input vary between basins in the Baltic Sea, depending on the type and volume of past and current industry in the coastal area (Korhonen et al., 2013; Salo et al., 2008; Sundqvist et al., 2009; Verta et al., 2007). On the other hand, atmospheric deposition mainly depends on the long-range transport of air emissions (Assefa et al., 2014b; Assefa et al., 2018; Sundqvist et al., 2010). Different pollution sources have different dioxin congener profiles with different toxic potencies, which are important to the toxicity to organisms.

The uptake of POPs by organisms from water depends on the bioavailability of POPs. In the water column, POPs (especially PCDD/Fs) are adsorbed to particulate organic carbon (POC) (Armitage et al., 2009). Thus, increasing organic carbon concentration decreases the bioavailability of POPs to organisms. Eutrophication, in turn, increases the phytoplankton biomass, which forms a major part of the sedimenting organic carbon in the Baltic Sea (Leipe et al., 2011). Consequently, high productivity in the system results in concentration dilution of the POPs (Skei et al., 2000), as well as increased sedimentation, which is the dominant removal process of PCDD/Fs from the food chain (Armitage et al., 2009).

Predator-prey relationships explain the contamination patterns and pathways: the levels of bioaccumulation (i.e., the increasing of concentration in an organism's tissue over time) and biomagnification (i.e., the increasing of concentration from one trophic level to another) depend on the species abundance and role in food webs (Walters et al., 2008). The predator-prey relationship of herring and salmon is the strongest in the northern Baltic Sea, while interaction with sprat (*Sprattus sprattus*) is more pronounced in the southern areas (Jacobson et al., 2018).

Since bioaccumulation is time-dependent, the growth and lipid content of individual fish, and the factors influencing them, are important determinants for dioxin concentration (van der Oost et al., 2003). Fishing, in turn, may have a spatiotemporal impact on mortality and size distribution in the fish population, which may affect the intra- and inter-specific competition as well as the growth of the individuals, and further the dioxin concentrations (Kiljunen et al., 2007; Peltonen et al., 2007). Thus, human exposure to dioxins in Baltic Sea fish depends not only on the amount of fish consumed, but also on the size of the fish and the fishing area. The sizes of herring and salmon in different parts of the Baltic Sea, assumed to be compliant or non-compliant with the maximum allowed dioxin level, are provided in the Commission Recommendation (EU) 2016/688, while national recommendations on consumption in Finland and Sweden are given based on the granted derogation. Furthermore, there are several area-specific factors affecting fish consumption, including availability of fish, as well as cultural fish eating habits in the different countries (Pihlajamäki et al., 2019).

Based on this framing, we identified five key categories associated with the dioxin risk formation (Fig. 1): A) Dioxin pollution and sediment concentrations; B) Environmental conditions; C) Food web dynamics; D) Dioxin concentrations in herring and salmon; and E) Human exposure to dioxins from herring and salmon.

For the analysis, the Baltic Sea was divided into four study areas (Fig. 2) utilizing the ICES sub-divisions (SDs) as follows: (1) the Arkona Sea (sub-division 24); (2) the Baltic Proper (SDs 25–29 (28.1 excluded)); (3) the Gulf of Bothnia (SDs 30–31); and (4) the Gulf of Finland (sub-division 32). The selected division is based on similarities in dioxin pollution and loading into the sea area and sediment (Wiberg et al., 2013), dioxin concentrations in the prey species of herring and salmon (Nfon et al., 2008; Peltonen et al., 2014; Vuorinen et al., 2012), physical and chemical growth conditions for marine and freshwater adapted organisms (Leppäranta and Myrberg, 2009), and anthropogenic pressures such as fishing (ICES, 2017a, 2017b, 2017c) and eutrophication (HELCOM, 2018).

2.2. Systematic literature search

The online databases Scopus and Web of Knowledge were used in a systematic literature search for peer-reviewed journal articles written in English, in autumn 2018. The literature review was conducted by using the ROSES protocol for systematic reviews (Haddaway et al., 2018a, 2018b). The objective was to track comparable variables of the identified categories, given the purpose of the study and the areal division selected.

Table 1 presents the search strings (title, abstract and keywords searched) specified for Scopus and Web of Knowledge, inclusion criteria (purpose), and spatially comparable measures found (result) under each category A–E. The search term “Baltic Sea” was used in all searches (e.g. sediment AND dioxin AND “Baltic Sea”). The years of interest in the systematic search was set to 2000–2017 in order to include the most recent knowledge on variables affecting the dioxin cycle. Specifications on the inclusion criteria and additional information on database-specific search properties and results are presented in the supplementary material (*Data tables*, Table S1).

2.3. Data preparation

The data found from the literature was heterogeneous, and the number of publications per each measure to be compared was small. When comparable and commensurable, the data values were averaged for the synthesis. To illustrate the level of variability, the standard deviation (σ), and minimum and maximum values are provided in the data tables, when possible. The coding for the specific variables selected (listed in Table 2) is presented in the supplementary material (*Additional material to methods and data - Data coding*).

For areal ranking, the values of the variables were indexed using proportional normalization. The study area having the worst status of a certain variable was scored 100 points. Here, the worst status is defined from the dioxin risk point of view; in other words, the condition that

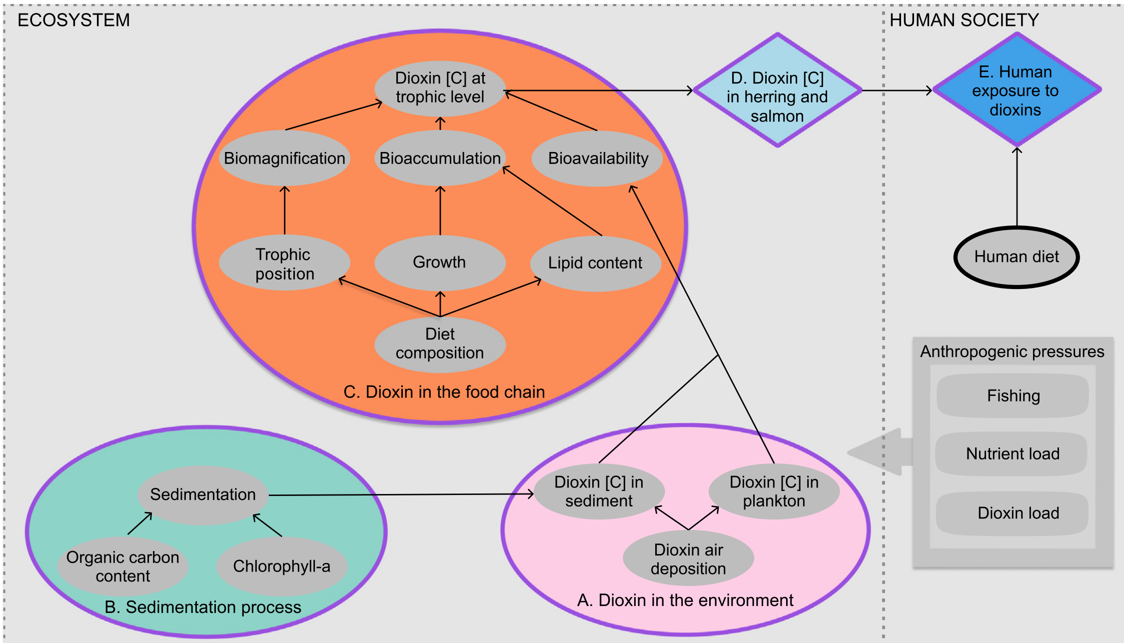

Fig. 1. Visualization of the analyzed system. The five steps of the causal chain leading to human dioxin risk are illustrated with purple-lined objects. Key variables (nodes) and causal mechanisms (arrows) affecting the dioxin accumulation and risks from Baltic herring and salmon are shown. Typical assessment endpoints are illustrated with diamond shapes. [C]: concentration.

most advances the dioxin accumulation to fish and/or human exposure to dioxins (see Table 2). The scores of the other areas were then calculated by proportioning their variable values to the value of the worst-case area. In the cases where the worst status is represented by the highest value (see Table 2), the following equation was used (Eq. (1)):

$$N_{j,k} = V_{j,k} / max_k * 100 \quad (1)$$

j and k are indices for study area and variable, respectively; N is the comparison index value; V is the actual data value; and max_k is the actual worst-case data value of the variable in question. When the worst case is represented by the minimum value (see Table 2), the following calculation was applied (Eq. (2)):

$$N_{j,k} = min_k / V_{j,k} * 100 \quad (2)$$

3. Results

The following section presents the literature search results, while the quantitative results in each category A–E are presented in sections 3.2.–3.6 and Supplementary material tables. The synthesis of the results is presented in Table 3.

3.1. Literature search results

The search strings yielded a total of 3954 records from Web of Knowledge and 866 results from Scopus (Fig. 3). Per category, the number of records in Web of Knowledge and Scopus was A) 281; 53 B) 396; 389C) 2000; 348 D) 724; 74 E) 553; 2, respectively. After removal of duplicates, the number of articles was 2693, of which titles and abstracts were screened simultaneously. A total of 81 full texts were screened, which resulted in 31 articles included from bibliographic databases (see Table S1). The most common reasons for exclusion were that the studies did not take place in the Baltic Sea, the spatial division was not stated explicitly enough for our areal division, they did not meet the criteria in a comparable data format, or the definition of the age of the specimen (in categories C and D) was not according to our criteria or not stated explicitly.

The search results from the Web of Knowledge and Scopus met the selection criteria for most of the categories. To supplement specific aspects in categories A and C, additional searches were conducted in Google Scholar for peer-reviewed and grey literature in English and Finnish (see 1.2. in the Supplementary material). A detailed list of records included in the final analysis is presented in Table S1. In total, these supplements resulted in the inclusion of 10 additional studies. Thus, taking into account the additional studies and four datasets (i.e. atmospheric pollution of PCDD/Fs and PCB-153 ([dataset] Gusev, 2018a; [dataset] Gusev, 2018b), herring weight-at-age ([dataset] ICES, 2011–2017), and herring and salmon catches ([dataset] ICES, 2017d)), the final number of studies was 46. The number of records per category were: A) 8, B) 7, C) 22, D) 7, and E) 2 (Fig. 3.).

Based on the search protocol and the identified materials for comparing the risk formation between the study areas, each category (A–E) was assigned a set of quantifiable variables. Part of the extracted data were available as such, whereas some were coded from the published results. The variables, their precise units of measurement, and method of whether they were extracted as meta-data or coded are listed in Table 2.

3.2. Dioxin pollution and concentration in sediments

The modelled deposition fluxes of PCDD/Fs (ng TEQ m^{-2} yr^{-1}) and PCB (ng PCB-153 m^{-2} yr^{-1}) area normalized to each study area are presented in Table S3. Both the depositions of PCDD/Fs and PCB-153 are the highest in the Arkona Sea (PCDD/Fs 1.38; PCB 289.40) and the lowest in the Gulf of Bothnia (PCDD/Fs 0.32; PCB-153 74.70). Moreover, high depositions are estimated in the Gulf of Finland (PCDD/Fs 1.12; PCB-153 177.90).

Dioxin concentrations in the sediments are presented in Table S4. The studies differed in their methodology and presentation of results. In the Gulf of Finland and Gulf of Bothnia, Korhonen et al. (2013) used sediment traps – devices that measure the quantity of sinking particulate matter – and presented the average concentrations of PCDD/Fs and dl-PCBs over the time the trap was deployed, whereas the other

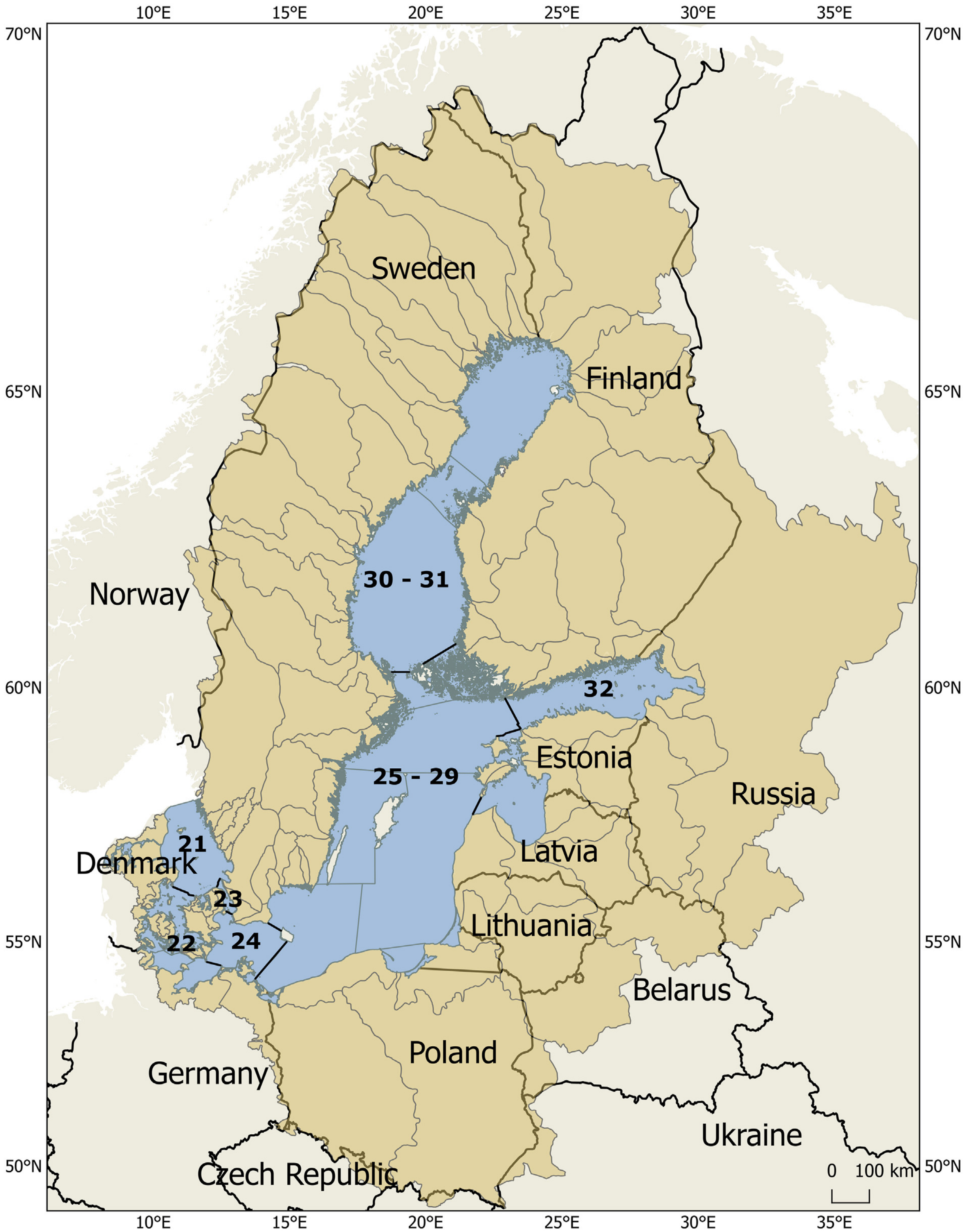

Fig. 2. Study area division in the Baltic Sea (light blue): (21–23 excluded), 24, the Arkona Sea; 25–29 (28.1 excluded), the Baltic Proper; 30–31, the Gulf of Bothnia; and 32, the Gulf of Finland. The light brown area represents the Baltic Sea catchment area. The map is created using Natural Earth data (www.naturalearthdata.com) combined with HELCOM data (http://metadata.helcom.fi) in QGIS.

studies used various corers and presented either average concentration of surface sediment or per core slice. Thus, only surface sediment values were used for the relative comparison of the areas. However, the sediment trap results by Korhonen et al. (2013) represent more recent concentrations compared to the other studies. In addition, the sediment concentration values of Dannenberger et al. (1997) were converted to WHO-TEQ$_{2005}$ format using the associated TEF-factors (Van den Berg et al., 2006).

The highest pelagic sediment concentrations of PCDD/Fs and dl-PCBs (pg WHO-TEQ$_{2005}$ g^{-1}) are measured from the Arkona Sea (PCDD/Fs: 7.69, $\sigma = 4.99$) and the Baltic Proper (dl-PCBs: 1.58, $\sigma = 1.79$), respectively. The concentration of PCDD/Fs in coastal sediments is clearly the

Table 1
Search strings (for title, abstract and keywords), inclusion criteria (Purpose) and the spatially comparable measures (Result) in different categories A–E. AND "Baltic Sea" was a common term in all of the searches. The search of the category E did not result in any inclusions.

Category	Search string	Purpose	Result
A) Dioxin pollution and sediment concentration	Sediment AND dioxin OR organochlorine	Measures for historical dioxin pollution	Measured sediment concentrations
B) Environmental conditions	"Particulate organic carbon" OR sedimentation OR "sediment accumulation rate"	Measures for comparing conditions for dioxin flux in the water column	Measured values of sediment accumulation rate and particulate organic carbon content
C) Food web dynamics	Herring OR salmon OR zooplankton OR benthic AND feed* OR diet OR isotope	Measures for comparing accumulation of dioxins to herring and salmon	Stomach contents and stable $\delta^{15}N$ isotope values
D) Dioxin concentration in herring and salmon	Herring OR salmon AND dioxin OR organochlorine	Measures for comparing dioxin concentration in herring and salmon	Dioxin concentrations along age and size
E) Human exposure to dioxin from herring and salmon	Herring OR salmon AND dioxin OR organochlorine AND "human exposure"	Measures for comparing human dioxin exposure from herring and salmon	–

highest in the Gulf of Finland (249.38, $\sigma = 199.88$) due to the influence of a former wood preservative factory (Korhonen et al., 2013). The concentration of coastal dl-PCBs is the highest in the Gulf of Bothnia (0.50, $\sigma = 0.62$).

In the synthesis, we compare the pelagic concentrations, because these values are better estimates of the general status of the area due to arbitrarily located land-based point sources on the coast. Pelagic sediments are also less prone to physical disturbance (e.g. water movement), hence reflecting a homogeneous and better comparable situation.

3.3. Environmental conditions

Table S5 lists the values compiled from the literature for the key environmental parameters defining the bioavailability and bioaccumulation of PCDD/Fs and PCBs in the Baltic Sea ecosystems, i.e. productivity (nutrients); it also includes sedimentation and POC deposition rates.

The average pelagic concentration ($\mu g\ l^{-1}$) of chlorophyll-a, which indicates the productivity of a system (Ferreira et al., 2010), is the highest in the Gulf of Finland (4.3) and the lowest in the Gulf of Bothnia (2.4) from the years 2011–2016. The Baltic Sea is prone to eutrophication, the excess of nutrients, due to long water retention times and stratification. In addition, the allochthonous loading to the Arkona Sea, the Baltic Proper, and the Gulf of Finland increases the eutrophication level notably.

Sedimentation, expressed as sediment accumulation rate ($g\ m^{-2}\ yr^{-1}$), is the highest in the Gulf of Bothnia (1620, $\sigma = 1843$), mainly due to a large amount of eroded sediment and river loads to the area (Mattila et al., 2006). The lowest sediment accumulation rate was estimated in the Arkona Sea (221, $\sigma = 128$). No studies of sediment accumulation rate in the Arkona Sea were available, and hence information from the Bornholm Basin and Bornholm Deep was used (Suplinska and Pietrzak-Flis, 2008; Winogradow and Pempkowiak, 2014).

The deposition rate of POC ($g\ C\ m^{-2}\ yr^{-1}$) is the highest in the Gulf of Finland (35, $\sigma = 0$), where large river input and water movement, combined with coastal topography, contribute to the deposition rates (Leipe et al., 2011). The lowest POC deposition rates are measured in the Gulf of Bothnia (20, $\sigma = 9$), and are partly explained by oxic surface sediment conditions, low productivity and strong currents (Leipe et al., 2011). No numerical data was available for the Arkona Sea, and hence the provided number may be an underestimation of the real value (see Kuss et al., 2006).

3.4. Food web dynamics

As illustrated in Fig. 2, bioaccumulation, biomagnification, and the resulting dioxin concentration at the trophic level are determined by trophic relationships, as well as lipid content and growth of the species. Diet-weighted biomagnification factor (BMF) for herring and salmon in different study areas is presented in Table S15. Data used in these

Table 2
The comparable variables, their units, the method of extraction (M: only summary data e.g. average available, C: detailed data used), and worst case explanations for variables (H: highest is the worst; L: lowest is the worst) under different categories: (A) Dioxin pollution and sediment concentrations; (B) Environmental conditions; (C) Food web dynamics; (D) Dioxin concentrations in herring and salmon; and E) Human exposure to dioxins from herring and salmon. In variable A1, no information on applied TEQ-protocol was given. BMF (C1–4): biomagnification factor. Adult herring: ≥ 5-year-old and/or ≥ 17 cm; adult salmon: ≥ 1-sea-year(s)-old and/or ≥ 60 cm. "Exposure by origin" (E1–4): burden of dioxin exposure of women 18–45 years from the catch landed from the study area (derived from herring and salmon consumption and catch data of Denmark, Estonia, Finland, and Sweden, combined with herring and salmon PCDD/F and dl-PCB concentrations in the study area).

Variable	Unit	Data	Worst case
A1: Atmospheric deposition of PCDD/Fs to pelagic water	$ng\ TEQ\ m^{-2}\ yr^{-1}$	C	H
A2: Atmospheric deposition of PCB-153 to pelagic water	$ng\ PCB\text{-}153\ m^{-2}\ yr^{-1}$	C	H
A3: Average pelagic concentration of PCDD/Fs in surface sediment	$pg\ WHO\text{-}TEQ\ g^{-1}$ d.w.	M	H
A4: Average pelagic concentration of dl-PCBs in surface sediment	$pg\ WHO\text{-}TEQ\ g^{-1}$ d.w.	M	H
B1: Average pelagic chlorophyll-a concentration in the system	$\mu g\ l^{-1}$	M	L
B2: Average sedimentation rate	$g\ m^{-2}\ yr^{-1}$	M	L
B3: Average particulate organic carbon deposition rate	$g\ C\ m^{-2}\ yr^{-1}$	M	L
C1: Herring diet-weighted BMF for PCDD/Fs	index	C	H
C2: Salmon diet-weighted BMF for PCDD/Fs	index	C	H
C3: Herring diet-weighted BMF for dl-PCBs	index	C	H
C4: Salmon diet-weighted BMF for dl-PCBs	index	C	H
C5: Average 5-year-old herring weight-at-age	g	M	L
D1: Average adult herring concentration of PCDD/Fs	$pg\ WHO\text{-}TEQ\ g^{-1}$	M	H
D2: Average adult salmon concentration of PCDD/Fs	$pg\ WHO\text{-}TEQ\ g^{-1}$	M	H
D3: Average adult herring concentration of dl-PCBs	$pg\ WHO\text{-}TEQ\ g^{-1}$	M	H
D4: Average adult salmon concentration of dl-PCBs	$pg\ WHO\text{-}TEQ\ g^{-1}$	M	H
E1: Herring exposure of PCDD/Fs by origin	$pg\ d^{-1}$	C	H
E2: Salmon exposure of PCDD/Fs by origin	$pg\ d^{-1}$	C	H
E3: Herring exposure of dl-PCBs by origin	$pg\ d^{-1}$	C	H
E4: Salmon exposure of dl-PCBs by origin	$pg\ d^{-1}$	C	H

Table 3
Comparison of variables (mean, min–max (where available)) and ranking of their index values in categories A–E in the study areas. For the variable units of measurement, see Table 2.

Variable	AS	BP	GoB	GoF	AS	BP	GoB	GoF
A1	$1.38_{(1.16-1.48)}$	$0.45_{(0.36-0.54)}$	$0.30_{(0.25-0.42)}$	$1.12_{(0.08-1.42)}$	100	32	21	79
A2	$289.4_{(271.9-314.1)}$	$87.3_{(81.2-91.3)}$	$74.7_{(68.5-80.2)}$	$177.9_{(163.5-191.0)}$	100	28	24	62
A3	$7.7_{(0.9-15.0)}$	$5.8_{(0.3-9.3)}$	$3.4_{(2.6-4.2)}$	$6.0_{(2.0-10.1)}$	100	75	44	78
A4	1.4	$1.6_{(0.3-2.8)}$	$0.6_{(0.6-0.6)}$	$0.3_{(0.2-0.3)}$	91	100	37	18
B1	2.6	3.2	2.4	4.3	92	75	100	56
B2	$221_{(68-471)}$	$409_{(60-6099)}$	$1620_{(110-6160)}$	$723_{(140-2490)}$	100	54	14	30
B3	25	$26_{(9-75)}$	$20_{(12-36)}$	35	79	76	100	57
C1	0.88	4.04	4.2	3.7	21	96	100	88
C2	1.76	1.33	0.52	0.4	100	74	32	23
C3	2.09	5.89	3.84	5.60	36	100	65	95
C4	2.64	2.21	0.99	0.8	100	80	39	30
C5	$153.9_{(136.0-165.0)}$	$42.8_{(21.4-77.7)}$	$29.9_{(27.0-32.0)}$	$23.6_{(19.0-27.6)}$	15	55	79	100
D1	$1.59_{(1.27-2.48)}$	$3.16_{(1.57-7.34)}$	$6.20_{(4.13-10.30)}$	$2.15_{(0.91-4.60)}$	26	51	100	35
D2	$3.84_{(2.58-5.09)}$	$4.30_{(2.03-9.27)}$	$3.86_{(1.56-5.86)}$	$4.20_{(3.57-4.79)}$	89	100	90	98
D3	$1.90_{(1.14-2.26)}$	$2.39_{(1.40-3.46)}$	$2.72_{(1.98-3.51)}$	$1.78_{(0.73-3.11)}$	70	88	100	65
D4	$8.06_{(5.41-10.71)}$	$6.97_{(3.20-14.20)}$	$5.52_{(2.86-6.80)}$	$7.13_{(5.13-8.15)}$	100	86	68	88
E1	0.3	4.2	3.9	0.5	6	100	93	12
E2	1.68E-03	2.84E-02	3.76E-01	2.97E-02	0	8	100	8
E3	0.3	3.2	1.7	0.4	10	100	54	13
E4	3.53E-03	4.61E-02	5.36E-01	5.04E-02	1	9	100	9

calculations are presented in Tables S6–S14. The diet-weighted BMF of PCDD/Fs estimated for herring was the highest in the Gulf of Bothnia (4.20), and in the Arkona Sea for salmon (1.76). The diet-weighted BMF of dl-PCBs for herring and salmon were the highest in the Baltic Proper (5.89) and the Arkona Sea (2.64), and the lowest in the Arkona Sea (2.09) and Gulf of Finland (0.80), respectively.

Herring weight-at-age (grams) is presented in Table S16 and illustrated in Fig. S2. An average value per study area was calculated for adult herring (5-year-old) from the years 2010–2016. For herring in the Baltic Proper and the Gulf of Finland, quarter-specific sampling data was available ([dataset] ICES, 2011–2017), from which the first quarter data was used. For the Arkona Sea and the Gulf of Bothnia, yearly average values ([dataset] ICES, 2011–2017) were used due to their availability. The highest and lowest herring weight-at-age were found in the Arkona Sea (153.9, $\sigma = 10.5$) and the Gulf of Finland (23.6, $\sigma = 3.2$), respectively.

3.5. Dioxin concentration in herring and salmon

Concentrations measured from pooled fish samples represent the most direct evidence of the dioxin quantities in the system. This is the most common monitoring method for estimating the risk of dioxins to human health in the sea area. The majority of the material in these studies is from the early 2000s, while the samples in the study of Hallikainen et al. (2011) were collected in 2009 and the study of Cantillana and Aune (2012) contained samples from 2001 to 2011.

Concentrations of PCDD/Fs and dl-PCBs in adult herring (≥ 17 cm and/or ≥ 5 years) and salmon (≥ 60 cm and/or ≥ 1 sea-year(s)) are presented in Table S18 and S20, respectively. The reported average concentrations in adult herring exceeded the EU limits for the selling of fish containing dioxins (EU, 2016) (PCDD/Fs: 3.5 pg g^{-1}; PCDD/Fs + dl-PCBs: 6.5 pg g^{-1}) only in the Gulf of Bothnia (PCDD/Fs: 6.20, $\sigma = 1.87$; dl-PCBs: 2.72, $\sigma = 0.51$). The reported average concentration of PCDD/Fs is the lowest in the Arkona Sea (1.59, $\sigma = 0.34$), whereas the lowest concentration of dl-PCBs is reported from the Gulf of Finland (1.78, $\sigma = 0.80$).

In contrast, the average concentrations of PCDD/Fs and dl-PCBs in salmon exceeded the EU limits in all study areas (Table S20), with the highest and lowest concentration of PCDD/Fs from the Baltic Proper (4.30, $\sigma = 2.84$) Arkona Sea (3.84, $\sigma = 1.77$), respectively. The average concentration of dl-PCBs in salmon was the highest in the Arkona Sea (8.06, $\sigma = 3.75$) and lowest in the Gulf of Bothnia (5.52, $\sigma = 1.39$).

3.6. Human exposure

The fishing intensity in the study areas of the studied countries (Denmark, Estonia, Finland, and Sweden) varies depending on the target fish abundance, as well as the capacity of the fishing fleet/traps and available quota in the area. This ultimately has an impact on the estimated burden of exposure (pg d^{-1}), BE_j. The BE_j for PCDD/Fs and dl-PCBs from herring and salmon are presented in Tables S24 and S25, respectively. Our estimation (Eq. S5) produces the highest average PCDD/Fs and dl-PCBs BE_j for the Baltic Proper herring (PCDD/Fs: 4.23; dl-PCBs: 3.20). For salmon, the highest average BE_j values for both compound types were found in the Gulf of Bothnia (PCDD/Fs: 0.38; dl-PCBs: 0.54).

3.7. Synthesis: comparison of the areas

The index values resulting from the variable normalization, compiled to evaluate differences in the regional risk formation, are presented in Table 3. The highest number of worst-status cases is shared between the Arkona Sea and the Gulf of Bothnia, which had seven variables with the maximum index value 100 and two and one variables over 90, respectively. The Arkona Sea holds the majority of the worst scores in the category related to dioxin pollution and sediment concentration of PCDD/Fs (variables A1–3), as well as the diet-weighted biomagnification of both PCDD/Fs and dl-PCBs for salmon (variables C2 and C4). In addition, the Arkona Sea scores worst in the average sedimentation rate (variable B2). In the Gulf of Bothnia, the worst cases relate to the low productivity (B1 – average pelagic chlorophyll-a concentration) and POC deposition (B3 – average POC deposition rate), the diet-weighted biomagnification of PCDD/Fs and concentrations of PCDD/Fs and dl-PCBs in herring (variables C1, D1 and D3), and human exposure to dioxins in herring (variables E1 and E3). The Baltic Proper has the worst status in five cases, but only one variable above 90. These concern the sediment concentration of dl-PCBs (variable A4), the diet-weighted biomagnification of dl-PCBs for herring and concentration of PCDD/Fs in salmon (variables C3 and D2), and human

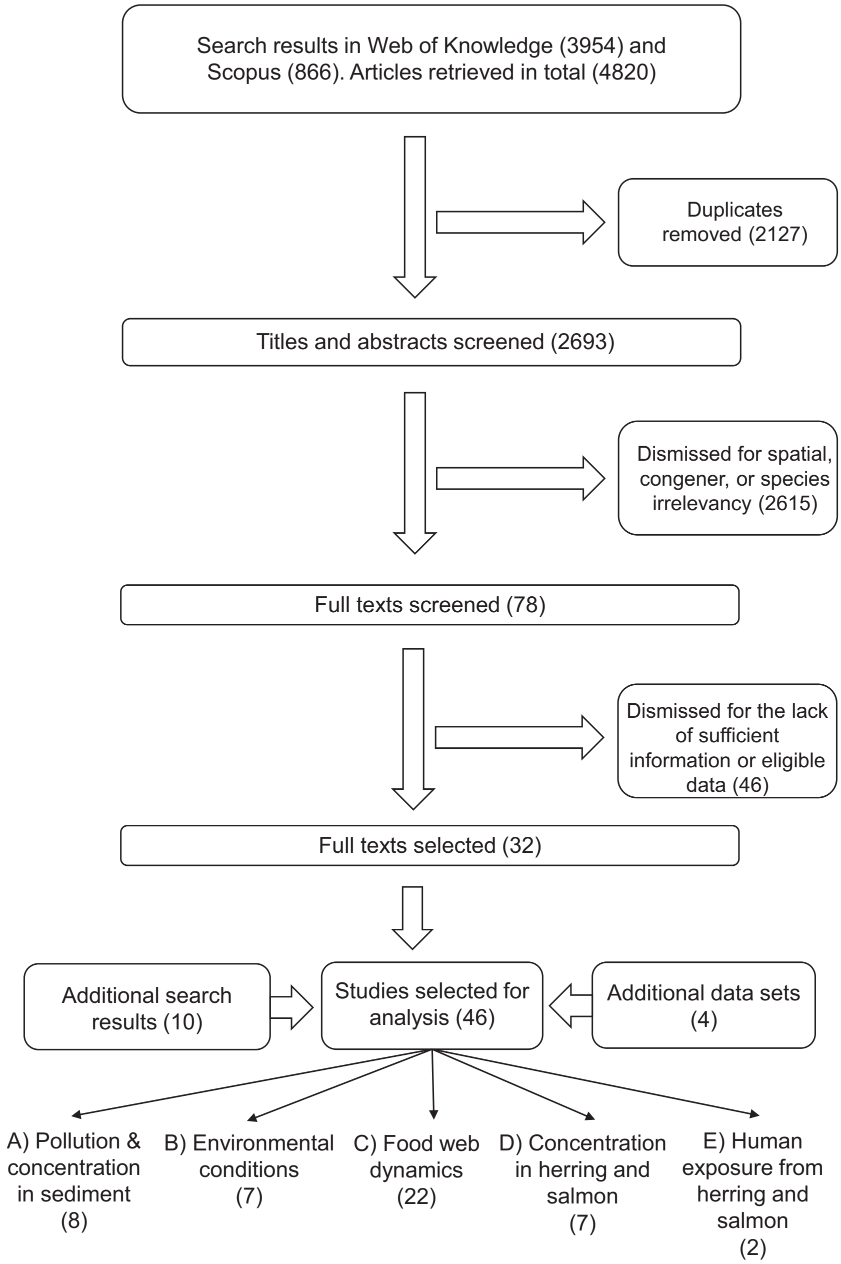

Fig. 3. Systematic search steps and resulting number of studies selected to the analysis categories A–E.

exposure to dioxins in herring (variables E1, E3). The Gulf of Finland has the only worst scores for the weight-at-age of herring (variable C5).

4. Discussion and conclusions

4.1. Spatial differences in the dioxin risk formation in the Baltic Sea

We have reviewed and synthesized existing knowledge concerning the transfer of dioxins from deposition to the ecosystem and further through the consumption of Baltic herring and salmon to humans, in order to compare the formation of dioxin risk in different parts of the Baltic Sea. The analysis demonstrates crucial differences between the four Baltic Sea basins concerning the contribution of different factors to the risk.

The spatial differences were most significant within category E – Human exposure to dioxins. The high variability is understandable, as it arises from multiple factors including differences in the dioxin concentrations in fish, origin of national catches, and fish consumption by humans. Together, the selling restrictions, national fish consumption recommendations, risk communication, and general perception of the risks and utilities related to Baltic fish eating all create variation in the way people consume fish, and consequently their exposure values (Pihlajamäki et al., 2019).

Category D, on the other hand, showed the smallest variation in concentrations of PCDD/Fs and dl-PCBs in salmon between study areas. This is reasonable, since the concentrations in salmon are partly a result of integrated accumulation over the areas due to feeding and spawning migrations. Population level dynamics are also important concentration determinants. For example, when the movement of salmon is explained by sprat migrations in the Baltic Proper or by more localized salmon preying on herring in the Gulf of Finland, and individual salmon length in relation to available prey length (Jacobson et al., 2018).

The study areas pose unique dioxin risk formation profiles. In the **Arkona Sea**, relatively high local and long-range atmospheric emissions have affected the higher concentrations of PCDD/Fs and PCBs in the sediment, compared to the more northern study areas (Assefa et al., 2014a;

Assefa et al., 2014b; Sobek et al., 2015). Since the sedimentation is clearly the lowest in relation to the other areas, we can assume that the removal of the compounds (especially the PCDD/Fs) from the water column to the sediment is the lowest in this area. However, the bioaccumulation and concentrations in herring in the Arkona Sea are relatively low due to the fastest individual growth in this area. The mixing of western Baltic spring spawning and central Baltic herring stocks is strongest in the Arkona Sea compared to the other study areas (Gröhsler et al., 2013), hence influencing the dioxin bioaccumulation characteristics. Exposure of young women in Denmark, Estonia, Finland, and Sweden (DK-EST-FI-SWE) to dioxins in herring and salmon caught from the Arkona Sea is minor compared to the more northern areas. This is mainly because only 3% and < 1% of the herring and salmon country-specific catches, respectively, originate from this area.

Similar to the Arkona Sea, the **Baltic Proper** is impacted by local and long-range atmospheric emissions more than the northern parts of the Baltic Sea (Assefa et al., 2014b). Due to the large area, there is an internal gradient: the deposition of PCDD/Fs and PCBs decreases towards the northern Baltic Proper. The dioxin concentrations in herring seem to be moderate in the Baltic Proper, while the human exposure values are the highest. All four countries (DK-EST-FI-SWE) fish in the Baltic Proper; in 2016, 38% of their joint herring catch originated from this area. Salmon, on the other hand, have high concentrations, yet the exposure values are low – only 6% of the joint catch is caught from this area.

In the **Gulf of Bothnia**, the atmospheric pollution of PCDD/Fs and PCBs, as well as concentration of PCDD/Fs in the sediment are the lowest among the study areas. Although the sedimentation rate in this area is the highest, due to low productivity, the sedimenting material is mostly inorganic, originating from the numerous rivers draining into the Gulf of Bothnia. On average, both herring and salmon dioxin concentrations exceed the maximum dioxin limits defined by the EU (2016) regulation.

The Gulf of Bothnia is the most important area for both herring and salmon fishing: 53% and 85% of the DK-EST-FI-SWE joint catches originate from this area, respectively. However, only Finland and Sweden fish in this area. Finland is the main user of Baltic herring: in 2016, they caught 56% of the total DK-EST-FI-SWE joint catch in the Baltic Sea, and 83% of the catch originating from the Gulf of Bothnia. Sweden uses the largest share of the Baltic salmon catch: in 2016, they used 50% of the total joint catch in the Baltic Sea, and 55% of the Finnish-Swedish catch in the Gulf of Bothnia. Accordingly, the exposure values from salmon were the highest in the Gulf of Bothnia. However, the highest exposure values from herring were found in the Baltic Proper. This reflects the fact that over 95% of the Finnish herring catch is not used in domestic food products, but rather in feed and fishmeal industry, as well as in exports (Setälä et al., 2016).

In the **Gulf of Finland**, the atmospheric deposition of both PCDD/Fs and PCBs, as well as the concentration of PCDD/Fs in the surface sediments were among the highest. Due to the small water surface area and volume, as well as the wide catchment area and numerous incoming rivers, the land-based allochtonous dioxin pollution and POC content assumingly contribute to the amount of sedimenting PCDD/Fs (Korhonen et al., 2013). Despite the high atmospheric deposition of PCBs, the pelagic sediment concentration of the compound type in our data was low.

The environmental conditions that decrease the bioavailability of dioxins (particularly PCDD/Fs), i.e. high sedimentation rate and high share of organic carbon in the sedimenting material, are on average the most optimal in the Gulf of Finland. The origin of organic matter influences the adsorption capacity of POPs, which is higher for autochthonous matter (Armitage et al., 2011). In the Gulf of Finland, both the terrestrial POC loading and primary production are considerable, which increase the general POC content and decrease the bioavailability of dioxins.

Our BMF calculations indicate that diet-weighted biomagnification of PCDD/Fs and dl-PCBs for herring were relatively high, with index scores of 88 and 95, respectively (Table 3). However, despite the lowest growth rate, dioxin concentrations in herring were found to be relatively low compared to the other study areas – especially in terms of PCDD/Fs. This might be connected to the environmental conditions (category B) that advance the storage and concentration of PCDD/Fs in the sediment. On the other hand, adult herring from the Gulf of Finland experience autumn migrations to the Baltic Proper and return to spawn in the spring (Parmanne, 1991). This reproductive behavior may partly explain the low concentrations. In the DK-EST-FI-SWE joint catches of Baltic herring and salmon, the shares of the Gulf of Finland (6% and 8% respectively) are minor, and consequently the calculated burden of exposure values of both species are among the smallest.

4.2. Accumulation of dioxins in Baltic herring and salmon

As part of this study, we evaluated the accumulation of PCDD/Fs, dl-PCBs and non dl-PCBs in Baltic herring and salmon by applying the theoretical BMF approach and combining it with the areal diets of herring and salmon (see section 2.1.3. in the Supplementary material). Our analysis is based only on the diets of adult specimens, and hence ignores the fact that the diet of both herring and salmon change along their life history (e.g. Jacobson et al., 2018; Peltonen et al., 2004). The diet-weighted BMF values summarize relevant information about the accumulation of dioxins in herring and salmon via their food, providing an indicator suitable for spatial comparison, which is the main focus of this evidence synthesis. For Baltic herring, comparison of the diet-based biomagnification of POPs has been conducted earlier, albeit with limited areal focus (Nfon et al., 2008; Strandberg et al., 1998; Burreau et al., 2006; Rolff et al., 1993; Broman et al., 1992). Moreover, none of these studies analyzed PCDD/Fs. When it comes to salmon, the issue has been addressed earlier in detail by Vuorinen et al. (2012) (and Burreau et al., 2006, to a lesser extent). In contrast to other studies, Vuorinen et al. (2012) did not include trophic levels in BMF calculus. The results of the diet-weighted BMF analysis are discussed in this section.

For herring, the diet-weighted BMF values (Table S15) indicate a stronger biomagnification rate of dl-PCBs compared to PCDD/Fs in all other areas but the Gulf of Bothnia. BMF values of dl-PCBs between herring and its key prey objects, cladoceran and copepods, in the Gulf of Bothnia are lower compared to the corresponding values of PCDD/Fs (Tables S11–S12; Fig. S1 C–D). One possible explanatory factor for the difference between PCDD/Fs and dl-PCBs is the higher transfer of POPs from the sediment – dominated by PCDD/Fs – to coastal organisms, compared to other parts of the Baltic Sea (Assefa et al., 2019). Studies in the Gulf of Bothnia have found evidence of the effect of polluted sediments on the coastal food webs (Sobek et al., 2014) and high bioaccumulation, particularly of PCDD/Fs, from sediment to herring (Assefa et al., 2019). Moreover, age-related accumulation of PCDD/Fs and dl-PCBs in herring is the most apparent in the Gulf of Bothnia (Vuorinen et al., 2012), mainly resulting from the slow growth of the fish in the area, but also due to above mentioned polluted sediments, influencing the high concentrations of PCDD/Fs and dl-PCBs in the prey species of herring (Table S6 and S7).

For salmon, diet-weighted BMF values for PCDD/Fs and dl-PCBs are higher in the south as compared to north. The BMF values to salmon (Tables S11 and S12; Fig. S1 A–B) suggest that salmon accumulates less PCDD/Fs and dl-PCBs from herring in the Gulf of Bothnia and the Gulf of Finland compared to the Baltic Proper and the Arkona Sea (i.e. less accumulation in areas where herring is the main prey). Three-spined stickleback bear the highest BMF value among the prey of salmon in almost all areas. However, its relative contribution diminishes in diet-weighted BMF due to its minor share in the diet of salmon. Bioaccumulation is generally higher with increasing lipid content of the prey, with an emphasis on dl-PCB accumulation (Vuorinen et al., 2012). In the Gulf of Finland and Gulf of Bothnia, the adult salmon specimens mostly feed on herring, which has distinctly lower fat content compared to sprat (see Table S8). Thus, the difference in diet-weighted BMF values may be due to adult salmon mainly feeding on fatty sprat in the Arkona Sea and the Baltic Proper (see Vuorinen et al., 2012; NB: the Arkona Sea was not included in the analysis). According to Kiljunen et al. (2008), herring in the Gulf of Finland have substantially lower energy contents compared to herring cohorts in other areas. Thus, salmon in the Gulf of Finland are required to feed more in order to meet their energy demands. On the other hand, in the Gulf of Bothnia, salmon preys, on average, older herring compared to the Baltic Proper (Mikkonen et al., 2011). Correspondingly, these factors cause elevated dioxin concentrations in the Gulf of Finland and Gulf of Bothnia salmon.

The study area-specific trophic level values result from system productivity, position of the organism in the food chain, as well as the variation in prey availability. Spatial and temporal food availability and energy demand impact the predator bioenergetics, isotope values, as well as the concentration of POPs (Kiljunen et al., 2008). Declining trends that have been observed in salmon isotope values reflect the changes in herring and sprat stock biomasses (Torniainen et al., 2014). In general, the δ^{15}N isotope values were the highest in the Gulf of Finland (Table S9), where the presence of cladoceran *Cercopagis pengoi* in the herring diet (Table S9) and high productivity (Table S5) partly impact the δ^{15}N isotope values and biomagnification. The explanatory power of trophic levels to explain the dioxin concentrations can be interpreted from the slope of the linear regression. Kiljunen et al. (2008) observed similar slopes in salmon studied in the Gulf of Finland, Gulf of Bothnia, and northern Baltic Proper, suggesting that salmon bioaccumulate dioxins rather similarly within the northern Baltic Sea area.

Our calculation of herring BMF and diet-weighted BMF involved simplifications, originating from data constrains and methodological variation among the diet studies included. Consequently, the regional differences in the (diet-weighted) BMF values for herring arise only from the relationship between lipid and dioxin concentrations of organisms, and partly from the variation in δ^{15}N isotope values. Excluding benthic species from the diet-weighted BMF analysis explicitly underestimates the regional magnitude of biomagnification: the lack of TEQ-based concentration data prevented valid comparison between areas. Moreover, the absence of benthic species in the diet of herring from the Gulf of Finland gives a misleading picture of the situation: a recent study by Kiljunen et al. (2020) highlights the importance of benthic energy sources to the upper level consumers in the Gulf of Finland food web. On the other hand, the abundance of benthic species such as amphipod *Monoporeia affinis* has declined in the Gulf of Bothnia (Kuosa et al., 2017), which impacts both the energy (Karlson et al., 2020; Kiljunen et al., 2020) and transfer of PCBs (Nfon et al., 2008) in the pelagic food web.

The BMF for salmon, in turn, is based on the weighted dietary differences between the study areas. Hence, the resulting values reflect the intra- and inter-specific competition, as well as the following stock sizes of its prey species (herring, sprat, and three-spined stickleback). The recent increase of three-spined stickleback in the diet of salmon could not be detected in our analysis, even though its share can occasionally be significant on local scales (Vuorinen et al., 2014). Indeed, Kiljunen et al. (2020) showed how three-spined stickleback dominate over herring in the Gulf of Finland and the northern Baltic Proper. Presumably, the spatial dietary variation is counterbalanced by the migratory behavior of salmon, resulting to rather similar dioxin concenrtations between study areas.

4.3. Management implications

The formation of dioxin-driven human health risks associated with fish species that are high in fat is a complex causal chain of phenomena; it starts with land-based emissions, continues through manifold complex ecosystem processes, and finally ends with the people who eat fish. Understanding how the different elements of this chain contribute to the risk in different parts of the Baltic Sea provides an interesting standpoint for the risk perception, both at the pan-Baltic and regional levels.

Improved knowledge about dioxin emission sources in different parts of the Baltic Sea can help in the design of different types of instruments to manage the emissions. In the southern Baltic Sea, long distance and regional atmospheric emissions from industrial, municipal, and domestic sources are the main contributors to the dioxin risk (Assefa et al., 2019; Wiberg et al., 2013; HELCOM, 2010). International regulations are essential in emission reduction; the regulations have resulted in, for example, up to 90% reduction in industrial emissions (Wiberg et al., 2013). On the contrary, the fraction of the domestic sources of all emissions has increased (Wiberg et al., 2013). For example, waste processing (e.g. disposal and incineration), as well as public electricity and heat production contribute to greater emissions of PCDD/Fs and PCBs, respectively, in the Baltic Sea catchment area compared to industrial processes (e.g. chemical and metal production) (EMEP, 2017). Here, economic incentives towards cleaner technology, material circulation, and efficient solutions, for heat production, for example, can be effective, together with the regional-level recommendations of HELCOM (2010). Information and education campaigns implemented by the municipalities could be a significant way to affect small-scale pollution sources, such as domestic and backyard burning (Wiberg et al., 2013).

For the polluted sediments, for instance in the Gulf of Bothnia and Gulf of Finland, the alteration of sediment geochemistry with sorbent amendments such as activated carbon, might be a local remedy (Ghosh et al., 2011). Activated carbon has reduced the concentrations and bioavailability of POPs in the pore water (see Patmont et al., 2015). In the coastal areas, sorption of PCDD/Fs and PCBs to sediment total organic carbon is weaker, while the transfer from sediment to bottom water is usually higher than in the offshore areas (Sobek et al., 2014). Coastal area sediments are under physical stress due to wave exposure and resuspension events, which does not allow equilibrium between pore water and organic carbon to be reached (Sobek et al., 2015). When contaminated sediments are located in an unstable environment, conventional methods, such as dredging and capping can be more suitable options than activated carbon (Gustavson et al., 2008).

In theory, dioxin concentration in fish can be regulated by harvesting the younger year classes or by increasing fishing mortality (MacKenzie et al., 2004; Peltonen et al., 2007; TemaNord, 2010). Such measures aim to decrease intraspecific competition, hence increasing growth rate. Targeting younger year classes would also result in catches consisting of smaller fish, complying with the maximum dioxin levels of the EU (Pihlajamäki et al., 2018). In practice, however, reducing dioxins by managing the fishery might be against the principles of ecosystem-based and sustainable fisheries management, as it might result in undesirable outcomes in the food web dynamics (Zhou et al., 2010). In addition, the benefits of fisheries management to decrease dioxin concentrations in fish may be relatively negligible, since the share of dioxins stored in biota is minor relative to the abiotic environment (Mattila and Verta, 2008). Selective fishing on an area-basis would imply a shift of fishing area from e.g. the Gulf of Bothnia to the Baltic Proper, to target fish with lower dioxin concentrations for human consumption. This could be possible in principle if fishers have quotas in different fishing areas, yet it might cause difficulties e.g. due to distances between homeports and landing sites.

Over the past 20 years, discussions have continued about the negative health effects caused by dioxin intake via fish, as well as the comparisons of these risks against the health benefits of consuming fish. Indeed, the debate is still lively, as new information accumulates and our risk perception is updated accordingly. Currently, the main instrument of the EU for reducing human exposure to dioxins is based on selling restrictions that are specific to the area of origin (EU, 2016). The decreasing emissions to and concentrations in the Baltic Sea ecosystem, together with the arguments about the health benefits overriding the potential risks, have raised the question of whether the management of the dioxin risk through eating recommendations – instead of selling restrictions – could be adopted all over the EU (Pihlajamäki et al., 2018). According to the Swedish recommendation, risk groups, i.e. children and women in the fertile age range, should not eat fatty Baltic fish more than two or three times a year, and other people should eat it once a week at the most (Livsmedelverket, 2018). The Finnish recommendation allows the risk groups to eat Baltic salmon and large Baltic herring (over 17 cm) up to once or twice a month, whereas restrictions for other consumers do not exist (Ruokavirasto, 2018). At the same time, the latest research results have demonstrated that our knowledge concerning the negative health effects of food-originating dioxin exposure may be still developing.

By reviewer's request, we applied data on herring and salmon consumption, origin of national catches, and total dioxin concentrations (PCDD/Fs + dl-PCBs) were applied to estimate the weekly dioxin intake of an average female (age: 18–45 years old; weight: 65 kg) in each country (Eq. S6), without any selling restrictions (i.e. the average concentration of the fish eaten was based on the proportional catch origins only). The results suggest that the EFSA TWI value would be exceeded by 70% and 20% in Estonia and Sweden, respectively (Table S26). Herring consumption among the risk group members in these countries (6.7 and 3.2 g d^{-1} per person, respectively) is considerably higher than in Finland and Denmark (1.2 and 0.9 g d^{-1}) (Table S21). This result could be used to consider, for example, if selling fish caught from a different Baltic Sea region could decrease the exposure of the risk groups in Estonia and Sweden. However, it is notable that these calculations are based on the dioxin concentrations in large, adult specimens only and hence the effects of the current dietary recommendations or selling restrictions to the size distribution of the fish eaten are not considered. Thus, the concentrations do not fully comply with country-specific, typical herring or salmon in the market basket either (see Pihlajamäki et al., 2019). Furthermore, due to continuous decreasing of dioxin concentrations in the environment and biota, our results may be an overestimation of the current dioxin concentrations in fish and subsequent weekly intake.

4.4. Limitations and development ideas

Ecotoxicological risk assessment studies have traditionally focused on analyzing either the concentrations of POPs in the environment, or the exposure of humans to those pollutants. The analysis here allowed us to understand the dioxin risk in a systemic manner, combining environmental, ecological and human-related factors at the regional level. Due to the complexity of the analyzed system and the small-scale spatial resolution, the availability of comparable data was fairly limited, hence applying statistical tests for validating the comparability was not meaningful. Nevertheless, this knowledge synthesis provides an indicative, comparable picture of the formation of the dioxin risk in the different Baltic Sea basins.

In the synthesis, we used averages over the observations reported for the areas as proxies for the status of each variable. However, many factors such as regional and seasonal variance, heterogeneous methods and sample sizes, as well as varying numbers of studies found for different areas, cause uncertainty in these calculations. Sampling points and method criteria in the different studies were chosen according to a specific focus, hence the picture of the situation in our study area is not necessarily the most comprehensive. The data in this study was gathered for a restricted timeline (2000–2017). However, both the dioxin pollution and concentrations in the Baltic Sea ecosystem have shown significant since the 1970s. Therefore, the selected timeline adequately represents the prevailing dioxin problem in the Baltic Sea during the first two decades in the 21st century.

The variable C5 (average 5-year-old herring weight-at-age) demonstrates that the size of herrings at a certain age vary remarkably between the study areas, as well as between the samples within each area (Table S16). As the concentration values under category D are measured from the pooled fish with the average minimum age of five years and/or average minimum length of 17 cm, the variability in the size of the fish used to calculate these concentrations is relatively high as well. Finally, acknowledging that the reported amounts of herring consumed by the responders of the consumption survey (2019[1]) are probably quite rough estimates, and also that the dioxin concentrations may include notable variation even in herring of the same age and size living within the same study area, the exposure estimates are unavoidably very uncertain.

To increase transparency of the results, we have provided the observational ranges, whenever available, in the tables of the article and the related support material. However, due to the varying number of studies included in the synthesis, the provided min and max intervals are not an explicit description of the within-area variation.

Recognizing the regional differences in risk formation may aid in directing the local focus in risk management, as well as provide valuable information for improved knowledge. The applicability of mechanistic contamination flux models parametrized with basin-specific variables could provide valuable information for management. Moreover, such methods would produce quantified knowledge of e.g. the responsiveness of the study areas to deposition reductions, or the effectivity of the current dioxin policy and regulations to protect the risk groups. In addition, probabilistic analysis methods such as Bayesian inference and Bayesian Networks (Nielsen and Jensen, 2009) could be used to analyze the epistemic uncertainty and comprehensively acknowledge it throughout the whole causal chain of the risk formation. For example, the population-level weekly intake estimates are associated with uncertainty arising from the randomness of the dioxin concentrations among the fish consumed, as well as the fish consumption and body weight of the individual consumers (see Fig. S3). The systemic causal presentation of Bayesian Networks would be a natural approach to describe the social-ecological interrelations, and also allow the optimal risk management measures under uncertainty to be identified (see e.g. Laurila-Pant et al., 2019; Pihlajamäki et al., 2020).

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CRediT authorship contribution statement

Lauri Nevalainen: Investigation, Systematic review, Conceptualization, Methodology, Data Curation Software, Writing - Original Draft. **Jouni Tuomisto:** Methodology, Writing - Review & Editing. **Päivi Haapasaari**: Conceptualization, Writing - Review & Editing. **Annukka Lehikoinen**: Supervision, Conceptualization, Methodology, Validation, Writing - Review & Editing.

Declaration of competing interest

None.

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[1] Survey was conducted in 2016 and data was available for use before publication http://en.opasnet.org/w/Goherr:_Fish_consumption_study

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